SPUTTER DEPOSITION AND CHARACTERIZATION OF METAL SANDWICHED INDIUM TIN OXIDE/SILICON FOR SOLAR CELL APPLICATION

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DEDICATION

To my loving and caring parents: Maryam Rabiu and Ishaq Aliyu. May Allah (SWA) continue to grant them more health with Iman, amen. I also dedicate it to my beloved wife Shamsiya Abdullahi Usman and my daughters: Maryam Kabir and Fatima Kabir respectively.

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ABSTRACT

For the past four decades, indium tin oxide (ITO) thin films have been widely used as transparent and conductive contact layer in several optoelectronic applications due to their high optical transmission and good electrical conductivity. Because of the rapid advancement of optoelectronic devices, new ITO design and structures are required to improve their performance for use in advanced applications. In this work, multilayer films structures of ITO/Al-Ag/ITO (IAAI) on n-type Si and ITO/Al-Cu/ITO (IACI) on p-type Si were prepared at room temperature by radio frequency (RF) and direct current (DC) magnetron sputtering technique. The microstructural, topological, morphological, optical and electrical properties of the prepared films were investigated after undergoing post-annealing treatment in a range of 300-600^{o}C in air. X-ray diffraction analysis of the as-deposited IAAI and IACI films show an amorphous structure with low intensity and broad diffraction Ag peak at (111) appearing on IAAI film indicating the presence of Ag film in a crystalline phase. The presence of In_2O_3 crystals peaks with Ag (111) and Al (200) for IAAI films and Cu (111) for IACI films were observed after post-annealing treatment indicating a crystalline improvement in the films. Topological analysis reveals an improved smooth surface topology and grain size with increasing post annealing temperature. The IAAI and IACI films showed better and improved microstructure at 500^{o}C and 600^{o}C respectively. The optical analysis shows a significant increase in transmittance with increasing annealing temperature. High optical transmittance of 91.01 % and 88.70 % for IAAI and IACI films annealed at 500^{o}C and 600^{o}C were obtained. The lowest sheet resistance of 3.81 Ω/sq by IAAI film annealed at 500^{o}C and 3.26 Ω/sq by IACI film annealed at 600^{o}C were obtained. Current-voltage characteristics analysis indicates a superior ohmic behaviour for IAAI films deposited on n-type Si and rectifying contacts on p-type Si for IACI films after post-annealing treatment making them an excellent IAAI/n-Si and IACI/p-Si contacts candidates for Si solar cells application.

ABSTRAK

Selama empat dekad yang lalu, filem tipis indium tin oksida (ITO) telah digunakan secara meluas sebagai lapisan sentuhan lutsinar dan konduktif dalam beberapa aplikasi optoelektronik kerana penghantaran optik yang tinggi dan kekonduksian elektrik yang baik. Oleh kerana kemajuan pesat peranti optoelektronik, reka bentuk dan struktur ITO baharu diperlukan untuk meningkatkan prestasi mereka untuk digunakan dalam aplikasi lanjutan. Dalam karya ini, pelbagai struktur filem ITO / Al-Ag / ITO (IAAI) pada Si jenis-n dan ITO / Al-Cu / ITO (IACI) pada Si jenis-p telah disediakan pada suhu bilik oleh teknik percikan magnetron frekuensi radio (RF) dan arus terus (DC). Ciri-ciri mikrostruktur, topologi, morfologi, optik dan elektrik filem-filem yang disediakan disiasat selepas menjalani rawatan penyepuhlindapan pasca dalam julat 300-600 °C. Analisis belauan sinar-X daripada filem-filem IAAI dan IACI yang dimendapkan menunjukkan struktur amorfus dengan puncak belauan Ag (111) yang lebar dan keamatan yang rendah muncul pada filem IAAI yang menunjukkan kehadiran filem Ag dalam fasa hablur. Kehadiran puncak bagi hablur In_2O_3 puncak dengan Ag (111) dan Al (200) untuk filem IAAI dan Cu (111) untuk filem IACI diperhatikan selepas penyepuhlindapan pasca yang menunjukkan peningkatan penghabluran dalam filem. Analisis topologi mendedahkan topologi permukaan licin dan saiz bijirin yang bertambah baik dengan peningkatan suhu pasca penyepuhlindapan. Analisis optik menunjukkan peningkatan ketara dalam transmisi dengan peningkatan suhu pasca penyepuhlindapan. Transmisi optik setinggi 91.01% dan 88.70% untuk filem IAAI dan IACI pada 500^0C dan 600^0C diperolehi. Rintangan setinggi terendah sebanyak 3.81 Ω / persegi oleh IAAI pada 500^0C dan 3.26 Ω/ persegi IACI filem pada 600^0C diperolehi. Pencirian arus-voltan mendedahkan tingkah laku ohmik yang terbaik untuk semua filem multi-lapisan yang didepositkan pada Si jenis-n dan kerintangan sentuhan pada Si jenis-p selepas rawatan pasca-penyepuhlindapan menjadikan IAAI / n-Si dan IACI / p-Si sebagai calon sesentuh terbaik untuk aplikasi sel suria.

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LIST OF SYMBOLS AND ABBREVIATIONS

ω	-	Angular frequency
Å	-	Armstrong's
β	-	Beta
σ	-	Conductivity
ε'	-	Dielectric constant
ε	-	Dielectric permittivity
ε_o	-	Dielectric permittivity of free space
°C	-	Degree celsius
e	-	Electron charge
υ	-	Frequency
μ	-	Micro
Ω	-	Ohms
%	-	Percentage
π	-	Pi
τ	-	Relaxation time
K	-	Restoring force
θ	-	Theta
d	-	Thickness of the wafer
Al	-	Alminium
AZO	-	Alminium zinc oxide
AFM	-	Atomic force microscopy
A	-	Area
Ar	-	Argon
R_b	-	Backside reflectance
cm	-	Centimeter
Cu	-	Copper
D	-	Diameter

EDX	-	Elemental dispersive x-ray
FESEM	-	Field emission scanning electron microscope
FOM	-	Figure of merit
FTO	-	Flourine doped tin oxide
4PP	-	Four-point probe
FWHM	-	Full width at half maximum
GZO	-	Gallium zinc oxide
g	-	Gram
Hz	-	Hertz
h	-	Hour
ITO	-	Indium tin oxide
IZO	-	Indium zinc oxide
IAAI	-	ITO/Al-Ag/ITO
IACI	-	ITO/Al-Cu/ITO
kg	-	kilogram
m	-	Meter
RMS	-	Root mean square
RT	-	Room temperature
RP	-	Rotary pump
R_{st}	-	Sheet resistance
Si	-	Silicon
Ag	-	Silver
Sq	-	Square
T	-	Temperature
R_{bT}	-	Total backside reflectance
TCC	-	Transparent conductive contact
t	-	Thickness
TCEs	-	Transparent conductive electrodes
2D	-	Two dimensional
3D	-	Three dimensional
TCO	-	Transparent conductive oxide
UV-Vis	-	Ultraviolet-visible spectrophotometer
XRD	-	X-ray diffraction

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CHAPTER 1

INTRODUCTION

1.1 Background to the study

Presently, transparent conducting oxides (TCO) thin films are attracting intense attention due to their high optical transmittance in the visible region, good electrical conductivity and various industrial applications. TCO films are important components for a wide variety of photosensitive electronic devices, acting as transparent and conductive window in solar cells, flat panel displays (FPD), light-emitting diodes (LED) touch screens and electrochromic devices (Ellmer, 2012; Granqvist & Hultake, 2002; Marikkannu *et al*., 2014; Meshram *et al*., 2015; Zhou *et al*., 2012). These materials have attracted a lot of interest, since after the first report of transparent conducting cadmium oxide films in 1907 (Bashir, 1998). The increasing demands and advancement of optical and optoelectronic devices such as portable and flexible electronics, displays, solar cells, multi-functional windows and not long ago the transistors are some of the reasons for this huge interest (Ho & Garcia, 2017; NCERT, 2014). Similarly, integration of different materials like metals, semiconductors, ceramics, plastics molecular and polymer organics into these devices have called for the provision of an improve TCO materials with required functional morphology, high performance and new processibility (David & Honoso, 2010).

High optical transmittance and infrared reflectance, and good electrical conductivity of TCO are the major factors that made the material attractive to researchers and industries (Guillén & Herrero, 2011). Over the years, new TCOs materials and their production process have been uncovering in the rush for technological advancement and market developments of the materials. The historical development of TCO materials are tabulated in Table 1.1. Some of the commonly discovered TCOs are indium tin oxide (ITO), zinc oxide (ZnO), fluorine tin oxide (FTO), titanium oxide (TiO_2), gallium zinc oxide (GZO), tin oxide (SnO_2), aluminium zinc oxide (AZO), gallium indium oxide (GIO) and antimony doped tin oxide (ATO). (Fortunato et al., 2008; Guillén & Herrero, 2011; Suzuki et al., 1996). Recently, thin metals films (silver Ag films), polymers, nanotubes composites, graphene, sulphides, nitrides and selenides are considered as transparent conductors (David & Honoso, 2010).

Table 1.1 TCO historical development (David & Honoso, 2010; Gordon, 2011)

Material	Process	Year
CdO	Thermally Oxidation	1907
SnO_2:Cl	Spray Pyrolysis	1947
SnO_2:Sb	Spray Pyrolysis	1947
SnO_2:F	Spray Pyrolysis	1951
In_2O_3:Sn	Spray Pyrolysis	1951
Cd-O	Sputtering	1952
In_2O_3:Sn	Sputtering	1955
In_2O_3:Sn	Spray Pyrolysis	1966
SnO_2:Sb	CVD	1967
Cd_2SnO_4	Sputtering	1974
ZnO: In	Spray Pyrolysis	1984
ZnO:Al	CVD	1992
ZnO: Ga	CVD	1992
$ZnSnO_3$	Sputtering	1994
$InGaZnO_4$	Sintering	1995
InGaZnO	PLD	2001
TiO_2:Nb	PLD	2005

The incorporation of TCO layer on silicon (Si) solar cell is a great discovery in the quest for an improved photovoltaic devices. These layers, when deposited on Si work as front window ohmic contact, antireflection layer, rectifying junction and passivation layers in an active way (Kumar *et al.*, 2015). They are also responsible for high absorption of energy in the absorbing part of solar cells, because of the high transmittance in the visible region. ITO as one of the leading TCO material has been largely considered by the researchers and industries due to its versatile structural and optoelectronic properties (Gulen et al., 2013; Herrero & Guille, 2006). This type of TCO possesses stable chemical characteristics and can be easily patterned using well suitable techniques for device fabrications (Zhu *et al*., 2000). The stability property of ITO films in several different environments is an advantage over the rest of TCO thin films materials. Intrinsic oxides such as zinc oxide (ZnO), aluminium zinc oxide (AZO) and gallium zinc oxide (GZO) thin films have been shown to have relatively lower stability in term of electrical and chemical effects than non-doped ITO thin films (Minami, 2008). Thus, TCO like AZO has some advantages for a large-area application like low cost, non-toxicity, environmental stability and material availability. Still obtaining a lower resistivity in that material requires a high annealing temperature than in ITO (Guillén & Herrero, 2011; Kim *et al*., 1999; Liu *et al*., 2005).

The $In_3O_3:SnO_2$ (ITO) is a highly degenerate n-type semiconductor material with high transmittance (> 80%) in the visible region, low electrical resistivity ($< 10^{-4}$ Ωcm) and high chemical stability (Guillén & Herrero, 2011; Laux *et al*., 1998; Wu & Lee, 2005; Yun & Kim, 2015). It has a direct wide bandgap energy (3.5-5.1 eV) which produces high transparency in the visible region of the spectrum (Paine, 2011; Yun et al., 2015). The ITO transparency to visible light is attributed to its large bandgap energy, while its conductivity is due to the arising intrinsic oxygen vacancies and extrinsic Sn^{4+} doping (Lee *et al*., 2012; Manavizadeh *et al*., 2009). Indium (In) metal, a rare and expensive metal constitutes the majority part of the ITO material. This factor plays a huge role in the final cost of any ITO/Si devices. Hence, getting an effective ITO films of reduced thickness is important. But, reducing the ITO films thickness to less than150 nm usually increases the resistivity of the film due to classical size effect (Girtan, 2012) which is an issue.

Furthermore, the technological demand of ITO thin films of good optoelectronics properties for application in advanced photosensitive devices has call for the exploration of new ITO thin films design and structures for effective performance (Meshram et al., 2015). Lately, ITO/metal/ITO multilayer structure has been discovered for efficient performance (Isiyaku et al., 2020a; Guillén & Herrero, 2011; Kim, 2009; Meshram *et al*., 2015; Park *et al*., 2016). This multilayer structure has drastically reduces the In metal consumption and is reported to have improved the electrical properties due to inclusion of thin metal films. The optical and electrical properties of the multilayer contacts largely depend on both ITO and metal films quality. Thin metal films of Ag, Al, Ni, Cu, Au, Pd, Cr,Pt, Mo, and Mg have been used

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